X-ray detection using a superconducting transition-edge sensor microcalorimeter with electrothermal feedback

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We have developed a new type of x-ray detector based on a superconducting transition-edge thermometer operated near 100 mK. A superconducting quantum interference device is used to measure the current through the thermometer, and negative electrothermal feedback is used to improve the energy resolution and shorten the thermal time constant. We have used a detector mounted on a scanning electron microscope to measure the energy of titanium $K\alpha$ (4.5 keV) fluorescence x rays with a resolution better than 14 eV full width at half-maximum. Using two other devices, we have measured an energy resolution for Joule heat pulses of 2.6 eV at 1 keV and 0.2 eV at 4 eV, the best reported for any calorimeter. An electrical noise equivalent power of 3×10^{-18} $\mathrm{W}/\sqrt{\mathrm{Hz}}$ was also measured, suggesting the use of these detectors as infrared bolometers. [S0003-6951(96)00839-X]

X-ray microcalorimeters are being developed by a number of groups[1–3] for spectroscopic applications in materials analysis and x-ray astronomy, due to their good energy resolution, wide acceptance angles, large energy bandwidth, high efficiency, and good spatial resolution. An x-ray microcalorimeter consists of an absorber to stop and thermalize incident x rays, a thermometer to measure the resulting temperature rise, and a weak thermal link to a heat sink to allow the detector to return to its equilibrium temperature. The first x-ray microcalorimeters used insulating or superconducting absorbers (for low heat capacity) and a semiconductor thermistor thermometer. After more than a decade of development, these detectors have achieved impressive energy resolutions of 7.3 eV full width at half-maximum (FWHM) operated at 60 mK.[4] Unfortunately, further improvement in the resolution of these devices is difficult because advances depend on a reduction in the already small heat capacity. Statistical variations in thermalization of the x-ray energy in insulating and superconducting absorbers also limit the resolution. Further, these devices are intrinsically slow, limiting their use in materials analysis.

In this letter we present a new type of x-ray microcalorimeter, the electrothermal feedback transition-edge sensor (ETF-TES) microcalorimeter, which has the potential for improved energy resolution while using absorbers with heat capacities up to two orders of magnitude larger than previous microcalorimeters. This generous heat capacity budget allows the use of large normal metal absorbers, providing more efficient x-ray thermalization and allowing faster detectors.

Negative electrothermal feedback results when a superconducting film is voltage-biased and the substrate is cooled to well below the superconducting transition. This feedback produces a self-biasing effect that causes the temperature of the film to remain within its transition region. As the film cools, its resistance drops, and the Joule heating (V^2/R) in the film increases. A stable equilibrium is established when the resistance is reduced to the point where the Joule heating matches the heat flowing to the substrate.[5]

Electrothermal feedback also reduces the thermal time constant of the sensor. When an x ray interacts with the absorber it is rapidly thermalized, raising the temperature of the absorber and the superconducting film. The rise in temperature increases the film resistance and decreases the Joule heating. This reduction in heating compensates for heat from the x-ray absorption with an effective time constant that is shorter than the intrinsic thermal time constant of the system. The effective time constant with electrothermal feedback is $\tau=\tau_0/(1+\alpha/n)$, where $\tau_0=C/G$ is the thermal time constant in the absence of electrothermal feedback (the heat capacity C of the thermometer and absorber divided by the thermal conductance G to the heat sink), $\alpha=d\ \log R/d\ \log T$ is the logarithmic sensitivity, a dimensionless measure of the sharpness of the superconducting transition, and $n=d\ \log P/d\ \log T$ is the logarithmic derivative of the bias power with respect to detector temperature. Since superconducting films can be fabricated with $\alpha\sim1000$, and n is typically 3–5, this response time can be shortened by more than two orders of magnitude.[5] When the effective time constant is short compared to the intrinsic time constant, the detector operates in the extreme electrothermal feedback regime, where substantially all of the heat is removed by a reduction in Joule heating rather than by an increase in the heat flow to the substrate. The x-ray energy deposited in the film is then the integral of the reduction in feedback Joule heating, or the bias voltage multiplied by the integral of the change in the bias current.

The fundamental limit of the energy resolution of the ETF-TES, including the Johnson noise of the film and thermodynamic temperature fluctuations between the detector and the heat bath, is[5] $\Delta E_{\mathrm{FWHM}}=2.35\sqrt{kT^2C\sqrt{8n/\alpha}}$, which is lower than the FWHM thermodynamic energy fluctuations in the detector by a factor of about $2.5/\sqrt{\alpha}$. Since α can be greater than 1000, this limit provides a generous heat capacity budget. Thus, while silicon thermistor microcalorimeters are restricted to insulating and superconducting absorbers with very small heat capacities, the ETF-TES microcalorim-

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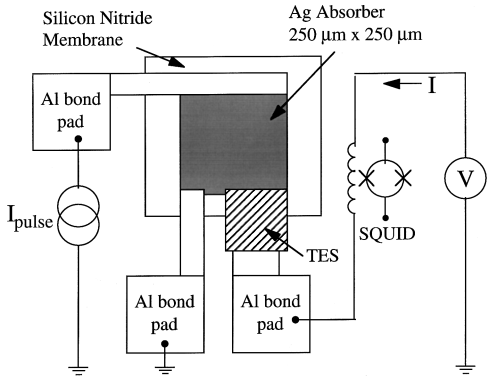

FIG. 1. The electrical bias circuit for the ETF-TES microcalorimeter and a diagram of the physical layout of detectors 1 and 2.

eter can use large normal-metal absorbers with heat capacities about two orders of magnitude larger. For example, for a gold absorber of dimensions 500 μm$\times$500 μm$\times$5 μm (which has a quantum efficiency of 99% for 6 keV x rays), with $\alpha=1000$ and $T=100$ mK, the fundamental limit on the resolution is 1.3 eV FWHM. Because a normal-metal absorber has no long-lived electron-hole pair or quasiparticle excitations, it has more efficient thermalization properties than an insulator or superconductor.[3] In addition, it can be designed with short diffusion times, making it possible to build a faster detector.

To improve the thermalization of the x rays in the absorber and to reduce the thermal coupling between the electrons in the absorber and the substrate, the devices described here were deposited on a freely suspended 0.5 μm thick silicon nitride membrane (Fig. 1). With this construction, most nonthermal phonons escaping from the absorber reflect off the silicon nitride/vacuum interface and reenter the normal metal where they are efficiently thermalized.[3]

The superconducting transition-edge sensor (TES) was fabricated as an aluminum/silver proximity bilayer. By varying the relative thicknesses of the aluminum ($T_c\sim 1$ K) and silver (normal metal) layers, we can tune the transition temperature of the bilayer to the desired range of 50–100 mK.[6] Using this technique, we are able to produce films with sharp transitions (transition width about 100 μK) that are reproducible to within 2 mK between different depositions. Other fabrication details were similar to those described elsewhere.[3,6]

The current through the ETF-TES microcalorimeter was measured using a series-array superconducting quantum interference device (SQUID) amplifier,[7] which has low noise and bandwidth greater than 1 MHz. The amplified SQUID current pulses were digitized before analysis. An electrical connection was also provided to allow the input of a fast heat pulse by dissipating Joule power in the absorber and thermometer (Fig. 1). This ability to inject heat pulses allowed us to calibrate the detector and study the energy resolution achievable in the absence of variations in thermalization and heat diffusion from an incident x ray.

We will present experimental results from three detectors (Table I). Detector 1 had a Ag absorber of area 250 μm$\times$250 μm and thickness 2 μm, a TES normal resistance of about 85 mΩ, and a T_c of about 85 mK. The detector was

TABLE I. Detector parameters and performance.

Detector	Absorber	E	ΔE_{FWHM}	τ_{eff}
1	250 μm$\times$250 μm$\times$ 2 μm Ag film	4.5 keV x ray	14 eV	480 μs
2	250 μm$\times$250 μm$\times$ 0.5 μm Ag film	1 keV heat pulse	2.6 eV	800 μs
3	100 μm$\times$70 μm$\times$ 50 nm Ag/Al bilayer	4 eV heat pulse	0.2 eV	8 μs

cooled in an adiabatic demagnetization refrigerator (ADR). Our compact, portable ADR uses two pills to cool from a 4 K liquid helium bath to about 50 mK without pumps,[8] and has a hold time of greater than 12 h at 100 mK. The detector is situated at the end of an ADR cold finger that is inserted into a commercial scanning electron microscope (SEM). The detector, which is 3 cm from the sample in the SEM, is in a separate vacuum space with a thin x-ray window. A titanium foil sample in the SEM was excited using an electron beam, producing fluorescent x rays that passed through the x-ray window and were absorbed in the detector. Pulses from the Ti $K\alpha$ (4.511 keV) and $K\beta$ (4.931 keV) lines were detected.

Pulse rise times, typically about 10 μs, were determined by the SQUID input and stray inductances divided by the bias resistance of the TES. Pulses had characteristic fall times of 480 μs with electrothermal feedback. When the operating temperature of the cold finger was within the superconducting transition (where there is little electrothermal feedback) we observed a characteristic fall time of about 5 ms. Electrothermal feedback, thus, shortened the pulse by about an order of magnitude. The magnitude of the feedback was limited by the critical current of the TES. In previous work,[9] pulse shortening by a factor of 100 was demonstrated using electrothermal feedback, and further optimization should yield similar results for our detectors. Even without increasing the existing amount of electrothermal feedback, it should be possible to reduce the pulse duration to less than 100 μs without loss of resolution by increasing the value of the thermal conductance between the detector and the heat sink.

The digitized x-ray pulses were analyzed using the optimal filter,[1] resulting in a sharp x-ray spectrum[10] [Fig. 2(a)]. Also shown is a spectrum obtained using a liquid-nitrogen-cooled commercial Si(Li) detector that was also installed on the SEM. The observed Ti $K\alpha$ line from the ETF-TES microcalorimeter is fit to the theoretical Ti $K\alpha 1$ and Ti $K\alpha 2$ line substructure using a least-squares method [Fig. 2(b)], determining a detector resolution of 13.9 eV FWHM, about an order of magnitude better than the commercial Si(Li) detector. The best energy resolution that has been reported for any calorimeter is 7.3 eV FWHM for the 5.9 keV Mn $K\alpha$ line using a silicon thermistor microcalorimeter.[4] Our result was achieved using an absorber with a heat capacity ten times larger, and with pulses ten times faster. Note that good energy resolution has also been achieved using superconductors–insulator–superconductor (SIS) tunnel junction detectors.[11,12]

The energy resolution of this detector was limited by SQUID noise that was measured to be 17 pA/$\sqrt{\mathrm{Hz}}$. We expect to improve our x-ray resolution by a factor of 2 by using

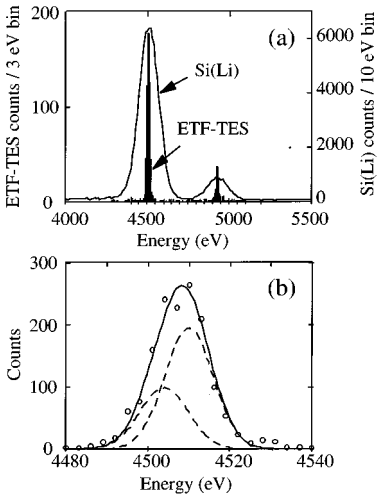

FIG. 2. Ti $K\alpha$ and $K\beta$ x-ray spectrum from an ETF-TES microcalorimeter connected to a SEM. (a) Both the ETF-TES microcalorimeter spectrum and a spectrum from a commercial, liquid-nitrogen cooled Si(Li) detector are shown. (b) The Ti $K\alpha$ microcalorimeter data (circles) is fit (line) to the theoretical Ti $K\alpha 1$ and Ti $K\alpha 2$ sublines broadened by 13.9 eV (dashes), determining an intrinsic detector resolution of 13.9 eV FWHM.

a SQUID with lower noise in the ADR. The other two detectors to be described here (detectors 2 and 3) were cooled in a dilution refrigerator that had a SQUID with a noise of 2 pA/$\sqrt{\text{Hz}}$. The dilution refrigerator was not, however, connected to our SEM to measure x-ray events.

Detector 2 had a Ag absorber of area 250 μm$\times$250 μm and thickness 0.5 μm, a TES normal resistance of about 1 Ω, and a T_c of about 80 mK. Heat pulses of 1 keV were injected into the absorber by joule heating. The resulting SQUID current pulses were analyzed using the optimal filter. Since this device was operated with less electrothermal feedback, pulses had longer time constants of 800 μs. Pulse energy resolution was 2.6 eV FWHM at 1 keV, an energy that caused temperature excursions to near the limit of the detector's linear response range.

Detector 3 was fabricated to determine the resolution attainable with small detector volumes. In this device, we used the Al/Ag bilayer itself as the absorber, with an area of 70 μm$\times$100 μm and 50 nm thickness. The normal resistance of the bilayer was about 1 Ω, and the T_c was about 85 mK. When heat pulses of energy 4 eV were injected, we measured a characteristic pulse time constant of 8 μs (due to the small absorber heat capacity), and an energy resolution of better than 0.2 eV FWHM. This result is the best energy resolution that has been reported for any calorimetric detector. Although the sensor would saturate at x-ray energies, this result suggests that the ETF-TES microcalorimeter can be used as high-efficiency, large-bandwidth optical photometer. Note that SIS tunnel junction detectors are currently used in this application.[13]

Finally, it has been suggested[14] that the ETF-TES would be useful as a fast infrared bolometer, since it can operate at speeds 100 times faster than the thermal time constant of the system. We discuss here the use of detector 3 for this application. The most important figures of merit for an infrared bolometer are its responsivity and noise equivalent power (NEP). For frequencies slower than the characteristic time constant of the system, the current responsivity of the ETF-TES is simply[14] $S_I=-1/V_{\text{bias}}$, where V_{bias} is the voltage bias of the sensor. We operated detector 3 with $V_{\text{bias}}=0.5$ μV, which yields a responsivity of -2×10^6 A/W. The current noise from the device was 6 pA/$\sqrt{\text{Hz}}$ and was white from the $1/f$ knee of the SQUID to the thermal roll-off frequency (about 20 kHz). From the current noise and responsivity, the electrical NEP was 3×10^{-18} W/$\sqrt{\text{Hz}}$. For this detector, the measured thermal conductance to the cold finger was $G=25$ pW/K. This NEP is consistent with the thermal fluctuation noise associated with this G, 2.2×10^{-18} W/$\sqrt{\text{Hz}}$, added in quadrature to the SQUID noise and Johnson noise from the bias circuit. Our calculated electrical NEP is comparable to that of the most sensitive direct detector of infrared radiation,[15] even though our thermal conductance is more than ten times larger.

Although the performance of these first ETF-TES x-ray microcalorimeters was far from the theoretical limit, our results are extremely promising. We have reported the first SEM x-ray spectrum taken with a calorimeter. The excellent 14 eV FWHM energy resolution for 4.5 keV Ti $K\alpha$ x rays is currently limited by SQUID noise in our ADR. Our heat pulse energy resolutions of 2.6 eV at 1 keV and 0.2 eV at 4 eV are the best that have been reported for any calorimeter. Finally, our electrical NEP of 3×10^{-18} W/$\sqrt{\text{Hz}}$ is one of the best results for any bolometer.

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